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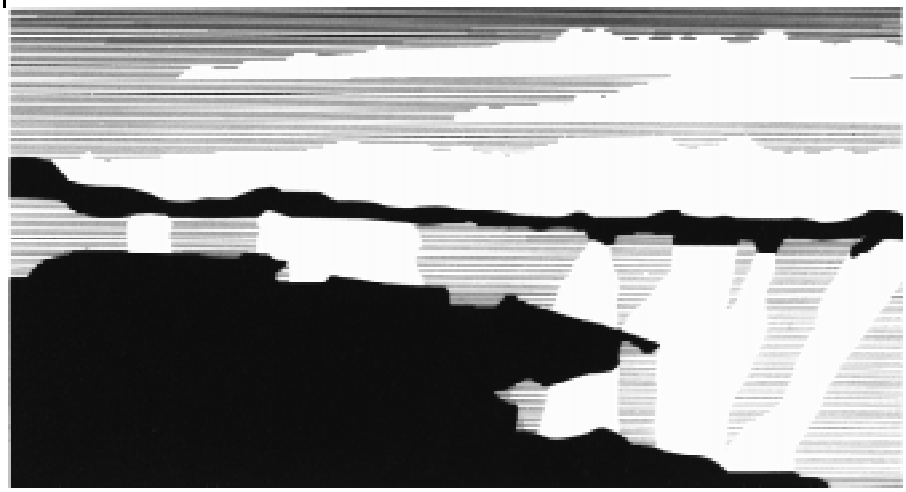

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Nondestructive Assay of Special Nuclear Material for Uranium Fuel-Fabrication Facilities*

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Abstract

A high-quality materials accounting system and effective international inspections in uranium fuel-fabrication facilities depend heavily upon accurate nondestructive assay measurements of the facility's nuclear materials. While item accounting can monitor a large portion of the facility inventory (fuel rods, assemblies, storage items), the contents of all such items and mass values for all bulk materials must be based on quantitative measurements. Weight measurements, combined with destructive analysis of process samples, can provide highly accurate quantitative information on well-characterized and uniform product materials. However, to cover the full range of process materials and to provide timely accountancy data on hard-to-measure items and rapid verification of previous measurements, radiation-based nondestructive assay (NDA) techniques play an important role. NDA for uranium fuel fabrication facilities relies on passive gamma spectroscopy for enrichment and U isotope mass values of medium-to-low-density samples and holdup deposits; it relies on active neutron techniques for U-235 mass values of high-density and heterogeneous samples. This paper will describe the basic radiation-based nondestructive assay techniques used to perform these measurements. We will also discuss the NDA measurement applications for international inspections of European fuel-fabrication facilities.

I. INTRODUCTION

Nuclear materials accounting and control (MC&A) in a nuclear facility depends upon the quantitative assessment of that facility's inventory of nuclear materials. This physical inventory must be based on accurate measurements of the nuclear materials, in all forms and locations in the plant. In a uranium fuel fabrication facility, nuclear material is received in bulk form and is processed into solid pellets of fuel material. These pellets are loaded into cladding (usually either stainless steel or zirconium) to form fuel rods or pins. The fuel rods are then gathered into assemblies, the basic unit of fuel for reactor cores. Feed material for a uranium fuel-fabrication facility is usually enriched uranium hexafluoride (UF_6); some facilities, however, begin their fuel fabrication process with UO_2 pellets imported from elsewhere. In Figure 1 is depicted the types of nuclear material found the U fuel-fabrication process, with emphasis on whether the materials exist in bulk form or as discreet items. Measurements are needed at all points at which nuclear material enters or leaves an accounting area, and this requires that many different measurement techniques be available to provide complete materials accounting data.

Determining the mass of uranium in nuclear materials requires a weight or volume measurement and a concentration measurement, which involves a variety of destructive or nondestructive techniques (Ref. 1). Destructive analysis is usually more accurate than nondestructive analysis, but it is slow and cannot be applied to many nuclear material forms. Destructive analysis cannot be applied to product materials such as fuel rods and assemblies, because of their high monetary value. Waste and scrap materials cannot be sampled for destructive analysis because of their heterogeneous nature. Nondestructive assay (NDA) was developed for such situations and is used frequently by safeguards inspectors and facility operators alike.

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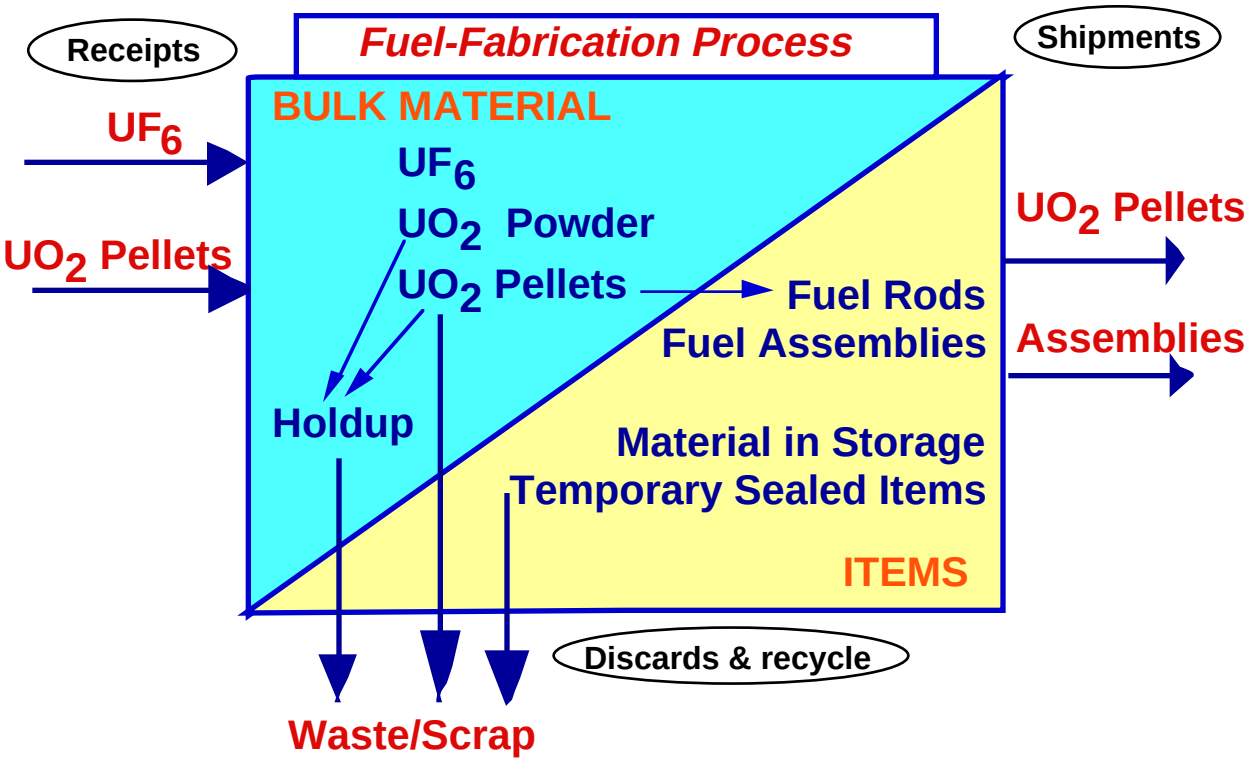

Figure 1. *Schematic of the flow of nuclear materials in a uranium fuel-fabrication facility. All of the processing that leads to the production of sealed fuel rods involves the movement and accounting for bulk materials. Once the rods are sealed, and their nuclear material content determined, they can be treated as items whose identification number is sufficient to determine how much nuclear material they contain. The diagram also points out nuclear material flow paths that include not only the process operations, but also the receipts, shipping, and waste pathways.*

II. NONDESTRUCTIVE ASSAY (NDA) TECHNIQUES

Weighing is by far the most precise nondestructive measurement of nuclear material, and most of the calculation of the final masses of well-characterized SNM samples is carried out on this basis. The weighing result is converted to quantity of uranium when the representative chemical and physical composition has been determined, usually by destructive analysis on analytical samples of the material. Difficulty arises, however, when matrix materials, variations in chemical composition, or impurities in the material are not well known and cannot be determined accurately by analytical techniques. Radiation-based nondestructive assay is then the preferred measurement approach and has its greatest value when the purity, past history, and chemical make-up of the material are not well-known. This approach is also widely used for rapid verification of previous measurements, regardless of their origin.

Radiation-based NDA is based on the measurement of gamma rays or neutrons, which penetrate sufficient material thicknesses to permit the assay of large items such as fuel assemblies and waste drums. Gamma rays readily penetrate low-density, low-atomic-number materials but are absorbed in high-density, high-atomic-number materials. Neutrons, on the other hand, penetrate large distances in materials like iron and lead but are severely attenuated by organic materials such as water and polyethylene. Therefore, the two radiations often provide complementary assay information.

NDA techniques can be active or passive. Passive techniques measure the spontaneously emitted radiation from nuclear materials, and active techniques use external sources to induce radiative emissions from the nuclear material. References 2 and 3 provide detailed discussions of NDA techniques.

A. Gamma-Ray Measurement of Bulk Material

Gamma rays interact with all matter, and this complicates nuclear material assay. The gamma rays born in nuclear materials do not all escape from the sample, and only a fraction of those that do escape interact with the detector. Uranium compounds are dense, high-atomic-number materials that scatter and absorb their own gamma radiations very effectively. Non-nuclear materials that are mixed with the uranium can also be efficient attenuators.

The most accurate attenuation-correction procedure, the transmission-corrected assay, uses an external gamma-ray source (transmission source) to measure the sample attenuation (see Fig. 2 and Ref. 4). The equation for this procedure is:

$$m = k\,A\,CF \qquad (1)$$

where m = mass of isotope measured
k = calibration constant
A = area (intensity) of assay gamma-ray peak
CF = attenuation correction factor.

The calibration constant is determined by measuring known reference standards. The attenuation correction factor is calculated from the measurement of the absorptive properties of the nuclear material sample being assayed. The key absorptive property measured is the fraction (T) of the incident radiation intensity (I_0) that passes through the full thickness (x) of the sample. This is determined by measuring the incident intensity from an external gamma-ray source, as well as the source intensity (I) transmitted by the sample. The fraction, T, is then determined by

$$T = I/I_0 = e^{-\mu\rho x} \qquad (2)$$

where μ = the sample material mass attenuation coefficient, determined by its chemical composition
ρ = the density of the sample material.

The transmission measurement, in effect, defines the quantity $\mu\rho x$ for the sample being assayed. In a far-field, transmission-corrected gamma-ray assay, the detector is far enough from the sample so that the radiation from the entire sample is determined in one measurement. The transmission measurement is performed through a small fraction of the sample volume; so for this measurement result to be applied to the full sample volume, the sample material must be uniform in composition and density. The extent to which this is not true (*e.g.*, lumpy or heterogeneous material) introduces bias in the assay. The primary gamma-rays used in uranium assays are given in table 1.

TABLE 1. Principal Uranium Gamma-Ray Signatures

Isotope	Gamma Ray Half-life (yr)	Gamma Ray Energy (keV)	Gamma Ray Emission Rate (g/s/g)
^{235}U	7.04×10^{8}	185.7	4.5×10^{4}
^{238}U	4.47×10^{9}	766.4	39
		1000.1	103

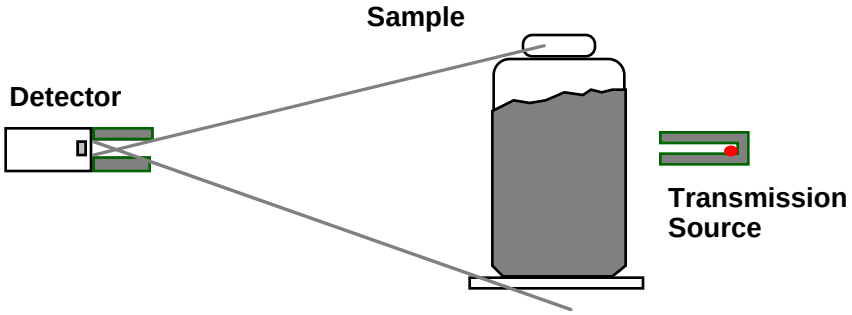

Figure 2. *Geometry for the transmission-corrected assay procedure in a far-field gamma-ray assay. The external source .for the transmission measurement (the transmission source) is centered on the axis of the gamma-ray detector, whose field of view encompasses the entire sample.*

The correction factor can be computed using numerical integration techniques and the $\mu\rho x$ for the sample material, or it can be estimated from approximate formulas such as:

$$CF = -\alpha \ln T / (1 - T^{\alpha}) \quad (3)$$

where α = 1 for a box, with flat, parallel sides
α = 0.825 for a cylinder.

The intensity of the 185.7-keV gamma radiation is used to determine the absolute quantity of ^{235}U in a sample by transmission-corrected gamma-ray assay methods (see Reference 5). The high absorption of the 185.7-keV gamma ray requires that such samples be very small, have low density, or be very thin deposits in plant equipment. Thick and/or dense samples will not allow the detector to view radiation from the full volume of the nuclear material and will result in a characterization of only the surface region of the sample. An alternate approach in such cases (especially for LEU) might be to measure the gamma intensity from the ^{238}U decays, which emit more penetrating gamma radiation at 766 and 1001 keV. Such an assay would allow determination of the amount of ^{238}U in the sample. In either case, these measurements must be combined with enrichment results for the same sample in order to establish the quantity of uranium in the item.

Portable gamma spectroscopy systems allow the operator to acquire spectra and analyze assay results in the field. (See Figure 3.) These capabilities are especially useful for internal inspections and audits of MC&A operations, as well as periodic measurements of in-plant holdup.

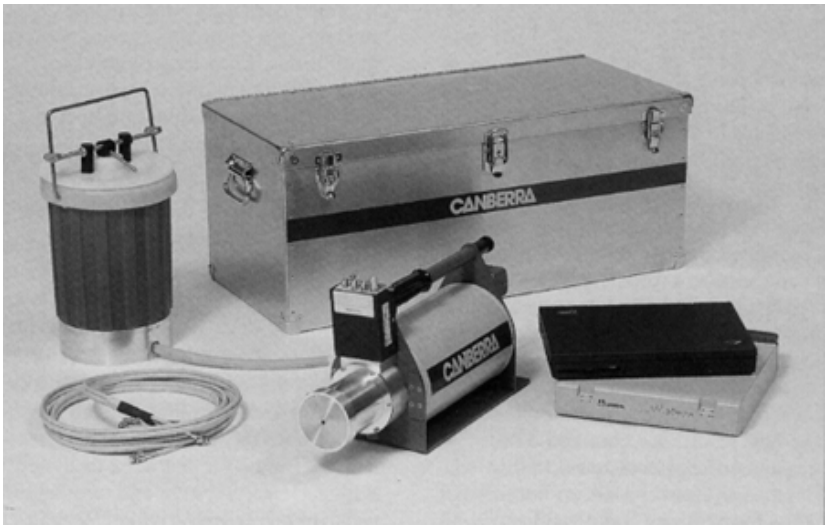

Figure 3. *Typical portable gamma spectroscopy system. The components include both low- and high-resolution (center of picture) detectors, data acquisition electronics and data analysis computer (right foreground). Accompanying software usually supports the full range of multichannel analyzer functions, as well as a variety of specialized assay options (enrichment measurement, mass assays, etc.).*

If samples are non-uniform, accurate assays may still be feasible if the sample is assayed in independent segments. The segmented gamma scanner (SGS, Ref. 5) uses the transmission-corrected technique, as shown in Fig. 4. It has a collimated detector and makes a vertical scan to perform transmission-corrected assays on cylindrical samples as a series of thin slices that may have different attenuations and nuclear material contents.

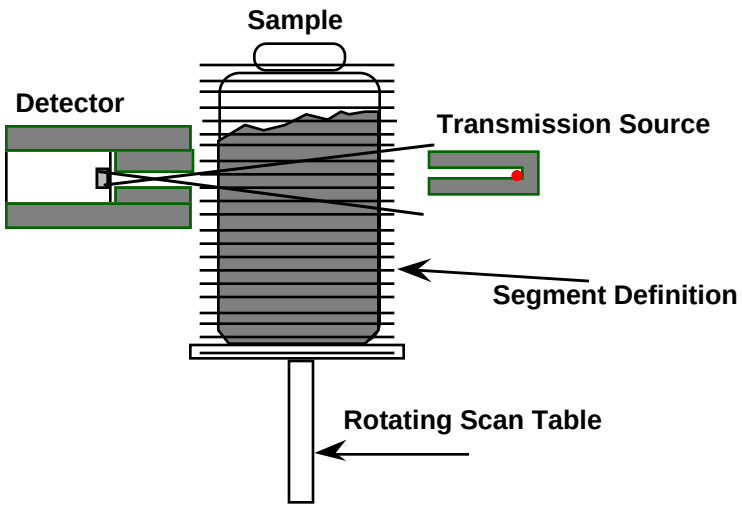

Figure 4. *Schematic of the measurement geometry for a segmented gamma scanner. The passive gamma emission and self-absorption correction are measured for each sample segment. The total sample assay is the sum of all of the transmission-corrected assays of the segments.*

B. Gamma-Ray Measurement of Enrichment

The fraction of uranium that is ^{235}U (*i.e.*, the ^{235}U enrichment) can be determined by destructive or nondestructive means. For high-quality accountability, it may be necessary to invest the time and money to determine the enrichment by destructive means (mass spectrometry) in order to achieve the smallest possible limit of error on the inventory result. This approach is practical with SNM that is both in bulk form and sufficiently homogeneous to allow acceptable sampling errors.

Nondestructive assay methods, involving gamma-ray spectroscopy, can be used to determine ^{235}U enrichment on a more timely basis. NDA methods are called for in cases where

- high measurement throughput is needed,
- measurement of a larger sample is required to obtain an average enrichment, because of sample inhomogeneities, or
- measured items are sealed or are products that cannot be opened or sampled.

If the uranium sample is large enough, then the 185.7-keV gamma rays originating from deep within the sample are completely absorbed and do not contribute to the gamma-ray intensity observed at the surface of the sample. Thus, with increasing sample thickness the 186-keV gamma-ray intensity at the surface reaches an equilibrium value, which is almost independent of the physical form of the sample. For pure uranium compounds this value is proportional to the ^{235}U enrichment of the sample, and generally only small corrections for chemical composition have to be applied. This is the “enrichment meter ” principle, and its application requires that the sample under assay is thick enough to be opaque for 186-keV gamma radiation (see Refs. 6-8). An instrument that uses this *enrichment-meter* technique is shown in Fig. 5.

Low-resolution measurements are vulnerable to gamma-ray interferences and yield enrichment results with relative accuracies of 1-5% (1σ). High-resolution measurements provide greater accuracy (0.1-1%), take somewhat longer, and require more expensive detectors.

Measurements with high energy resolution are recommended for

- UF_6 cylinders (where interferences can be encountered from material build-up on the inner surface of the cylinders, and the thick walls of the cylinders can cause scattering effects that cannot be corrected for),
- uniform bulk material for which high accuracy is desired,
- situations where container thickness or material attributes change a great deal, or
- situations when ^{232}U or ^{236}U concentration is high (above 0.1 ppm).

Low-resolution measurements are recommended for

- uniform bulk material for which 1-3% accuracy is adequate
- verification of enrichment on UO_2 pellets, powder, rods, and assemblies, or
- measurement of materials in similar containers.

Calibration for enrichment measurements involves the measurement of accurately characterized standards - usually composed of UO_2 powder. Enrichment measurements of other uranium compounds involves corrections to the measurement data. Other physical forms of the material (bulk quantities of sintered oxide pellets, scrap of varying composition, etc.) also require corrections to a calibration based on oxide standards. NDA enrichment measurement of individual fuel pellets, however, is also straightforward (See Refs. 6-8).

Figure 5*. Measurement of ^{235}U enrichment in cans of uranium oxide with a collimated NaI (low-resolution) scintillation detector. The portable multichannel analyzer on the right has 4096 channels, is battery operated, and guides the operator through the enrichment assay.*

C. Active Neutron-Based Measurements

If SNM samples are large, high-density, or heterogeneous, then the accuracy of gamma-ray assays is undermined by large, highly uncertain photon absorption effects. The less penetrating neutron radiation from fissions can therefore serve as an assay signal that is not as vulnerable to absorption by high-density materials.

Spontaneous neutron emission rates from uranium samples are too low to make passive neutron assays practical except for the most massive samples. Neutron-based assays of uranium materials must therefore involve active stimulation of fissions in the ^{235}U in the sample, followed by detection of either the prompt coincidence neutrons from the induced fissions or the delayed (singles) neutrons from the fission fragments. Both measurement approaches determine the quantity of the fissile isotope ^{235}U, and enrichment results on the same samples would be required to determine total uranium in the sample.

Active neutron coincidence counting is a particularly effective assay method for determination of ^{235}U in heterogeneous samples, in view of the high penetrability of neutrons (see Reference 9). Measurement of UO_2 (powder, bulk pellets, fuel rods, or fuel assemblies) by this method can provide the ^{235}U content with accuracies of 2-5%. This technique is not recommended for UF_6 measurements, largely because of the high singles neutron background from the fluorine matrix, but also because the measurement geometry for coincidence counting of large UF_6 cylinders is quite awkward.

For higher fissile mass sensitivity (*e.g.*, characterization of low-level waste), delayed neutron counting is the preferred method (see References 10 and 11). Instruments that implement this technique (so-called “shufflers”) employ a ^{252}Cf source to induce fissions in the sample and then (after the Cf source is quickly placed behind

shielding) count the delayed neutrons following the induced fissions. Sensitivities down to milligram quantities of fissile isotope are possible. For 1-kg or few-liter sample volumes, an accuracy of 0.5 to 5% is possible. For larger samples (e.g., 200-liter drums), accuracies of 1-10% are more likely.

1. Active-Well Coincidence Counter (AWCC)

The active-well coincidence counter counts the emission rate of prompt neutrons from induced fissions in the sample. The counter surrounds the sample with ^{3}He neutron detectors and irradiates the sample with neutrons from AmLi sources installed above and below the sample in the counter end caps (See Fig. 6). The analysis electronics measures the coincidence neutron count rate, which is proportional to the amount of fissionable material (^{235}U) in the sample. Coincidence counting differentiates the fission signal from the undesired neutron counts from the random AmLi interrogation source and from room background.

***Figure 6 (a)**. The AWCC, showing a typical assay sample. The coincidence electronics and analysis computer are on the bench behind the counter.*

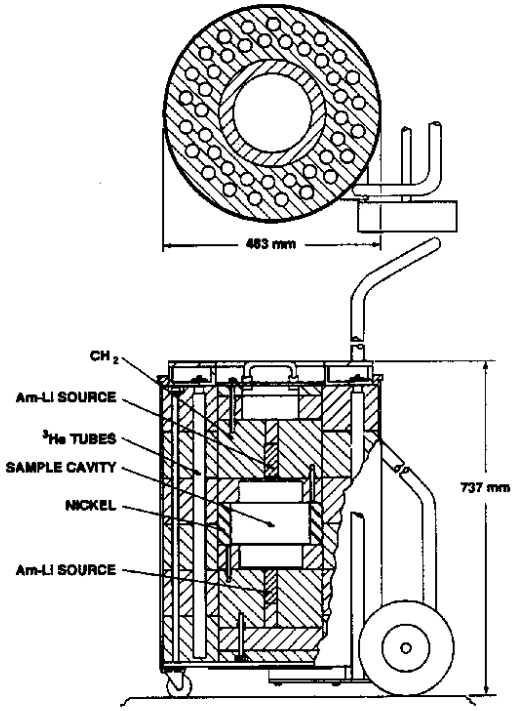

***Figure 6 (b).** Line drawing of the AWCC, showing placement of sample chamber, detector tubes, and the end caps that contain the AmLi interrogating sources.*

The AWCC is intrinsically less sensitive than a passive neutron coincidence counter, because the coincidence count rate from induced fissions is accompanied by significant (background) neutron counts from the interrogating sources that are integral to the counter. When the AWCC is operated with a Cd liner in the sample chamber, the interrogating neutrons are largely epithermal, and the fissile sensitivity is correspondingly reduced (the so-called "fast mode" of operation). With the Cd liner removed, thermal neutrons are free to interact with both the sample and the ^{3}He detectors, which significantly increases the response of the system and therefore its sensitivity (the so-called "thermal mode" of operation). The sensitivity of the AWCC (*i.e.*, the minimum detectable quantity of ^{235}U) is defined as 3 times the background σ. In the thermal mode, the AWCC sensitivity in 1000-sec counting times is approximately 1 g ^{235}U; in the fast mode, this sensitivity is approximately 25 g ^{235}U. The AWCC is best suited for high-mass, highly enriched uranium samples and should not be used for low-^{235}U-mass samples, except for well-defined samples in the thermal mode. (See Ref. 9)

2. The Uranium Neutron Coincidence Collar (UNCC)

For safeguards purposes, it is of high interest for inspectors to measure full fuel assemblies, because they constitute the output product from the plant and the input to the reactor facilities. Enriched uranium is often transferred from one installation or country to another in the form of fuel assemblies.

The Uranium Neutron Coincidence Collar (UNCC) uses an adaptation of the AWCC design to assay fissile content of fresh fuel. The fuel assembly is surrounded on three sides with ^{3}He neutron detectors, and the interrogating AmLi source is encased in moderating material on the fourth side of the assembly (see Figure 7). When no interrogation sources are present, the passive neutron coincidence rate gives a measure of the ^{238}U through the spontaneous fission reactions.

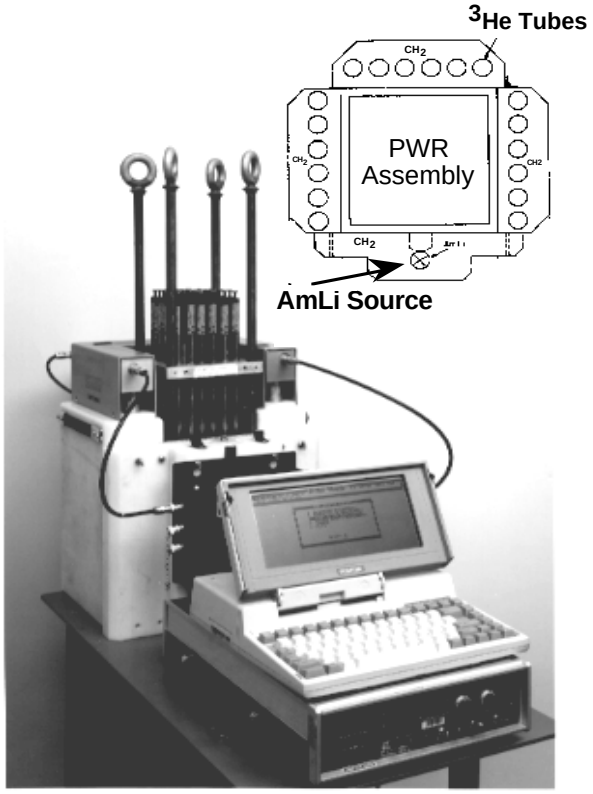

Figure 7. *Neutron Coincidence Collar shown measuring a PWR fuel assembly. This instrument uses an AmLi source to induce fissions in ^{235}U. The inset shows a cross section of the arrangement of detectors and interrogating source.*

The UNCC has been applied to measurements of boiling-water-reactor (BWR) and pressurized-water-reactor (PWR) fuel and to other types of fuel assemblies for accountability, criticality control, and safeguards purposes. Statistical precisions for 1000-sec assays vary from 0.6% to 0.9% (1σ), depending on the type of assembly. For longer counting periods, the ultimate precision was about 0.1% for repeated runs with a fixed geometry. Relative variations in the loading of an assembly as small as 1.9% can be detected in a measurement time of 1000 sec. The UNCC is in wide use for inspection applications.

3. Neutron interrogation, followed by delayed neutron counting (^{252}Cf Shuffler)

A shuffler (Refs. 11-13) performs active neutron assays for fissile materials in uranium (or any other fissile material) by repeatedly irradiating the item with neutrons from a ^{252}Cf source and quickly withdrawing the source so that delayed neutrons from the induced fissions can be counted (see Figure 8). (Delayed neutrons are emitted by fission fragments seconds to minutes after a fission.) Typical assay times range from 10 min. to 16 min., including a 3 or 4 min. background count and 20 to 35 "shuffles" of the ^{252}Cf. Materials assayed in shufflers have ranged from milligram quantities of uranium waste through kilogram quantities of purified uranium. The shuffler measurements play an important MC&A function by accounting for the SNM in scrap and waste. They also can provide valuable material control function by screening waste streams to assure that SNM is present at waste levels and that larger quantities of SNM are not being diverted out a pathway that should normally handle only small quantities of SNM.

The items assayed in shufflers are usually very diverse. All types and amounts of fissile materials may be encountered, and the packaging matrices may be equally important. For shuffler measurements of diverse items in a physical inventory, the materials to be assayed should be placed into categories (stratified), where each category exhibits similar physical and chemical properties and may share the same standards.

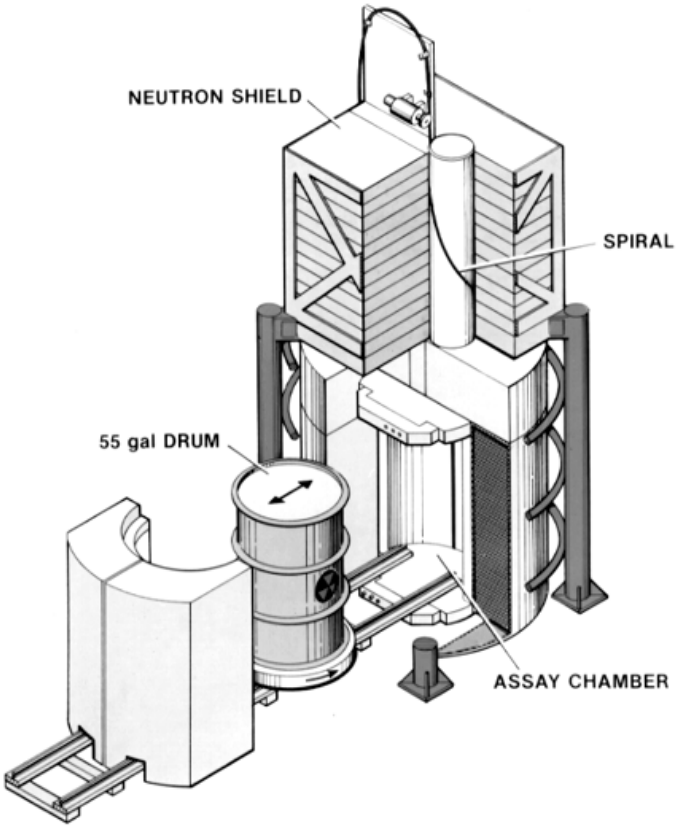

Fig. 8. *^{252}Cf shuffler for assay of 200-L drums of SNM-bearing materials. The interrogating ^{252}Cf source is stored in massive shielding in the top of the instrument and moves down into the assay chamber through a spiral channel. The assay chamber at the bottom of the instrument surrounds the sample with ^{3}He tubes to detect the delayed neutrons from the induced fissions.*

4. Rod Scanners

The SNM content of fuel rods is usually established by weighing as the rods are loaded with the fuel pellets. From that point on, the SNM content of the sealed rods is identified by its label (bar-code, engraved number, etc.). If subsequent measurements are performed on the rods, it is usually either for verification or for quality control of the SNM content of the rod. Rod scanners can perform these measurements by moving the rod past an intense neutron source, which induces fissions in the SNM. As the rod moves down from the neutron source, it moves past a neutron or gamma-ray detector, and the delayed neutron or gamma radiation level is measured, thus providing a measure of the fissile profile of the rod, which (when normalized to standard rods) can verify its SNM content. In addition, the fissile profile can also verify the pellet-to-pellet uniformity and the enrichment profile in rods that have variable enrichments along their length (See Fig. 9). Typical measurement times are 30 seconds for each rod, and measurement precisions are usually better than 1%.

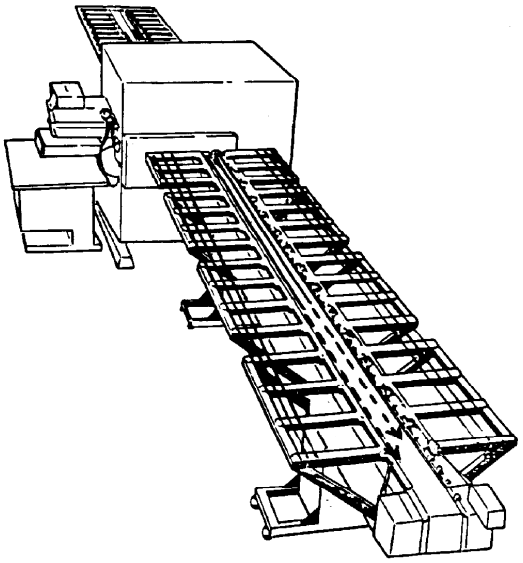

Figure 9 *: Active uranium fuel-rod scanner. Rods move past the shielded neutron source (in the rear of the picture) and then past a gamma or neutron detector. The induced activity is a measure of the fissile content of the rod at the location being measured.*

III. CALIBRATION AND MEASUREMENT CONTROL

A. Calibration and Standards

The nondestructive assay (NDA) of special nuclear materials (SNM) involves the measurement of the radiation emitted by samples of the material to determine the amount of material present, its isotopic composition, or both. These measurements must be carefully calibrated by establishing the response of the measurement instrument to known quantities of nuclear material, by applying the assay technique in question to thoroughly characterized samples that contain well-known quantities of the nuclear material. These samples are calibration standards, prepared from working reference materials. Their SNM values are measured using techniques that establish traceability of these values to the national measurement system. Certified reference material is available for gamma-ray enrichment measurements from the New Brunswick Laboratory (SRM 969) in the US and from the Central Bureau of Nuclear Measurement in Europe. See References 14 and 15 for more details on the preparation of calibration standards for various NDA techniques.

B. Measurement Control

In a high-quality NDA measurement program, the validity of the instrument calibrations and the overall quality of the measurement systems must be periodically verified through a conscientious measurement control program. This program begins with the fabrication of well-characterized, traceable standards, followed by procedures to assure that the standards and the measurement techniques retain their high quality. Maintenance of reliable measurement system requires

- calibration and use of well-characterized working NDA standards
- careful documentation of the traceability of those working standards to the national system of measurements, and
- regular diagnostic procedures on the instrument setup, measurement environment, data content, and measurement results (which may not require tracebale standards).
- a procedure for training operators, verifying continued operator competence
- documentation of all procedures, findings, and measurement results.
- detailed documentation of the precision and accuracy of each measurement system, both to demonstrate its continued quality and to provide input to the determination of the overall limit of error of the material balance.

IV. APPLICATIONS OF NDA MEASUREMENTS

A. Physical Inventory Taking

The physical inventory in a facility is based on measured values for all quantities of nuclear material. Even though, at inventory time, many items may already possess measured values for special nuclear material (SNM) content and verifiable integrity (through seals/TIDs), there will be many items that will have to be measured during the inventory-taking exercise. These are items that

- have been created in preparation for the inventory (*e.g.,* from clean-out operations)
- exhibit broken seals and whose contents must be re-verified, and
- constitute materials held up in process equipment and that have not been measured
- are sealed but are selected for verification measurements.

To the maximum extent possible, nuclear material items to be measured should contain SNM whose physical, chemical, and isotopic compositions are homogeneous and well-known. This allows the use of straightforward NDA techniques with maximum precision and accuracy. Any items that contain heterogeneous or poorly characterized SNM should be separated from the well-characterized items and measured by more complex and possibly intrusive means or with less accuracy.

An additional consideration is the cost-effectiveness of the measurement options in a plant. The least expensive measurement approaches are the gamma-ray techniques. To be effective, these techniques require specific sample attributes — low density, small samples size, and sample uniformity are the most important. Inventory items should be created with these requirements in mind to afford the maximum possible opportunity for gamma-ray assays where NDA is needed. Neutron-based NDA techniques require costly instrumentation, and so items that would require such measurements should be as small in number as possible. Finally, holdup measurements, even though they involve gamma-ray techniques, are often costly because of the large amount of labor required to make difficult measurements over many assay locations. Accordingly, every effort should be made to minimize holdup measurement requirements during inventory operations by thorough cleanouts and by well-documented holdup estimates from process histories.

B. Measurement of In-Plant SNM Holdup

Nuclear material left behind after the cleanout of the process, is deposited in various pieces of equipment in a variety of measurement geometries. In addition, deposit thicknesses and amounts and types of intervening materials (between deposit and detector) are often poorly known (or altogether unknown), contributing to large measurement uncertainties for assays of such deposits. The measurement of in-plant SNM holdup is both an art and a science and requires a great deal of experience and a practical sense of proportion on the part of the measurer to accommodate these large uncertainties and still generate useful measurement results. It is usually necessary to make some simplifying assumptions about each measurement and apply a few standard measurement geometries to the assay analysis in order to obtain a quantitative result (Refs. 16 and 17). Measurements of the same holdup deposit using several different geometry assumptions and from several different directions can reveal the magnitude of the appropriate overall uncertainty of the assay, which will usually exceed the uncertainty associated with only counting statistics.

For thin uranium holdup deposits, measurement of the 186-keV gamma ray from ^{235}U decay is recommended. The assay will determine grams of ^{235}U, which must be combined with a separate enrichment determination (either from NDA or from known process history) to obtain grams of uranium in the deposit. For thick holdup deposits, or deposits in thick-walled vessels, measurement of the 1001-keV gamma radiation from ^{238}U decay is preferable. The assay will determine grams of ^{238}U, which must also rely on a separate enrichment measurement to determine total uranium. In both cases, it is usually necessary to determine a correction to the measurement result for the gamma-ray absorption by the holdup deposit and by the intervening materials between the deposit and the detector. In many cases, the measurement geometry does not make it practical to use an external transmission source. In such cases, the measurer must make reasonable estimates of the attributes of the deposit (density, thickness, distribution) and calculate a correction factor using formulas such as that given in equation (3). If past history and clean-out results at the deposit site are available, this information will help achieve better accuracy in subsequent holdup measurements at the same location. If there are many holdup measurement sites that are repeatedly measured, then their physical attributes should be kept on a data base and the location labeled (perhaps

with a bar code) to facilitate rapid retrieval of information about the measurement site in subsequent measurement campaigns. (See Figure 10.)

Figure 10. *Portable measurement of nuclear material holdup in a ventilation duct. The measurement location has been permanently marked with a bar-code label. The operator is reading the bar-code label to identify the measurement site. He will then perform a gamma-ray assay at that site, using the low-resolution detector in his left hand. The multichannel analyzer electronics is strapped to his belt on his right side. [Note, picture is for demonstration purposes under contamination-free conditions; actual measurements of this type are performed with protective clothing.]*

In addition to measuring the quantity of nuclear material in holdup deposits, it is also possible in some cases to estimate its magnitude from historical process data. This approach is especially cost-effective and is therefore recommended if the quantity of the estimated holdup and its uncertainty are known to have negligible impact on the limit of error of the material balance.

C. Scrap and Waste Measurements

Assays of poorly-characterized items (whose gamma absorption, neutron moderation, or neutron production characteristics are poorly known) or assays of holdup deposits (whose measurement geometries are often highly uncertain) yield results with characteristically large assay uncertainties. In view of this, it is of paramount importance to minimize the amount of SNM in the inventory that is in these undesirable categories. In this way, the assays that must still be done on these types of materials will contribute as little as possible to the total limit of error on the material balance. Accordingly, prior to an inventory exercise, the process should be cleaned out as thoroughly as possible, and the SNM obtained from the cleanout should be stratified and packaged in easily measured containers for careful assays by methods outlined in the previous sections.

The remaining materials in the process and uncharacterized materials from cleanouts can then be measured by whatever methods are appropriate. Containers of scrap and other heterogeneous materials may be best measured by active neutron coincidence counting, to minimize the influence of unknown absorption effects on assay accuracy. Active coincidence counting of heterogeneous materials often shows accuracies 2 to 3 times worse than measurements on homogeneous materials. If the effect on the limit of error of the material balance is small from such samples, NDA using the 1001-keV gamma radiation may be adequate, and it is certainly a cheaper, faster, and easier measurement to perform.

1. Stratification:

As scrap items are created (during the routine production process or during preparations for an inventory), they should be carefully stratified by grouping together items that have similar known SNM characteristics and similar radiation-transport properties, so that they can be measured with similar calibration and analysis techniques. This practice improves the quality of the MC&A measurements by greatly reducing one major source of measurement uncertainty: unanticipated variations in sample characteristics. Stratification also improves the efficiency with which measurement and other materials management resources are applied to the inventory process, by focusing specific resources only where they are needed and where they are most effective.

2. Measurement of Scrap:

Items that contain SNM that is sub-standard in some way but is of sufficient quantity to merit recycling are defined as scrap. Some examples of scrap items are:

- rejected or broken pellets
- oxide that is out of specification
- damaged fuel rods
- bulk material recovered from a process upset (*e.g.*, UO_2 powder).

When scrap items are created from sources such as these, they should be stratified in such a way that past analysis data (combined with the weight of the items) can be used to assign an SNM quantity to the item. An example would be to stratify scrap by chemical composition and enrichment. Then the weight data can be supplemented by known enrichments and element factors to establish the quantity of uranium and of ^{235}U in the item. Once the items are so characterized, they should be sealed, so that no further measurement is necessary until they are recycled through the facility.

3. Measurement of Waste:

If items are created that contain a mixture of SNM attributes, and are of small enough quantity that it is not cost-effective to recycle them, they are characterized as waste. Some examples of waste items are:

- floor or glove box sweepings from process cleanout
- contaminated low-density materials (e.g., rags, anti-contamination clothing)
- high-density contaminated items (e.g., furnace bricks, pieces of discarded equipment)
- rejected or damaged product materials with a mixture of SNM attributes (e.g., chemical compositions, enrichments).

Low-density materials can usually be packaged in containers and assayed by transmission-corrected gamma-ray techniques (*e.g.*, SGS). Higher-density waste with mixed SNM attributes may require either high-energy gamma techniques (gamma assay using the 1001-keV radiation from ^{238}U decay) or neutron assay techniques (AWCC or Shuffler). The neutron techniques involve very costly instrumentation, so gamma-ray techniques should be employed whenever possible. This suggests that every attempt be made to create the waste items so that they will be assayable by gamma-ray means (low-density or small containers). SNM content of some difficult-to-measure waste items (e.g., furnace bricks or retort liners) might be estimated using holdup measurement techniques on selected samples of such items. It is desirable that these estimates have minimal impact on the material balance and its limit of error, since they will have large measurement uncertainties. This is possible to the extent that these items comprise a small fraction of the total inventory.

V. MEASUREMENT APPLICATIONS FOR INTERNATIONAL INSPECTIONS

Nondestructive assay measurements are a popular method for safeguards inspectors to verify operator-declared SNM values. The EURATOM inspectorate addresses the European facilities and complements the coverage of nuclear facilities by the IAEA inspections.

A. Unattended Measurement System (UMS)

A medium-term objective of the EURATOM Safeguards Directorate is to implement up to 100% verification of the output of all major LEU fuel fabrication plants by unattended measurement. To achieve this the JRC Sa-Ve-Tech unit has developed an unattended measurement system (UMS), for fresh LEU fuel assemblies. The system is an integration of a shift register based uranium neutron coincidence collar (UNCC) and optical character recognition to provide automatic identification of the assembly being measured (see Fig. 11). The system is designed

to provide unattended service for months without inspector intervention. The only intervention required of the facility operator is that of inserting and removing fuel assemblies. All activities of the measurements and identification system including loading and unloading of the AmLi neutron source are pre-programmed for automatic operation. The inspector can program the UMS to measure in different positions of the assembly and to make a scan measurement of the whole assembly. The results are stored in a protected database to await the next visit of the inspector. The software system controlling mechanical automation, data storage and evaluation as well as the user interface are implemented under Windows NT 3.4 using Visual C++ and connected to the database via ODBXC (Refs. 18 and 19).

The chief requirements of the system are:

- The facility operator has no involvement in the safeguards measurement other than to load the fuel assembly into the measurement station and unload it once the measurement is completed.
- The UMS is made with a neutron coincidence collar and permits automated measurement in passive mode and in thermal- or fast-active mode
- The fuel assembly identification number is read by a video system which is integrated into the measurement station.
- The mechanical components of the UMS include different detector heads which can be mounted to accommodate PWR or BWR assemblies, and software-based operation allows for measurement of different types of fuel assemblies in the same device.

The UMS is equipped with sophisticated software which

- allows the inspector to configure the automated measurement strategy, analyze measurement results and generate inspection reports,
- can, if required, make real-time evaluation of measurement results,
- allows the inspector to carry out measurements manually when he is present (hands-on-mode),
- monitors its own performance for maintenance purposes,
- protects itself against tampering, and
- can, if required, communicate remotely with an inspector via modem.

Figure 11*: The UMS performs a vertical scan of a fresh fuel assembly to obtain total fissile content of the assembly.*

Calibrations, using the EURATOM Performance Laboratory (PERLA) reference standards, and pilot tests have been performed at the Joint Research Center (JRC) Ispra. Field tests of the UMS system using BWR fuel assemblies have been performed at the Fabbricazione Nucleare facility in Bosco Marengo. The whole testing procedure has demonstrated

- reliability of the mechanical operation in automatic unattended mode,
- reliability of the optical character recognition under facility conditions,
- effectiveness of the calibration using FN assembly measurements and measurements on mock assemblies constructed from LEU pin samples from the PERLA laboratory, and
- interpretation of scanning profile and evaluation of its effectiveness for coverage of diversion strategies.

The UMS is made for use with the existing commercially available neutron collar designs (the Canberra JCC-72 for BWR assemblies and the JCC-73 for PWR assemblies) as well as for use with the next generation of neutron collars. Measurement precisions for the UMS are the same as those achieved by the uranium neutron coincidence collar (UNCC). The second generation collars, developed by the Sa-Ve-Tech unit of the JRC Ispra, will have a double row of ^{3}He detectors for increased efficiency. The fuel assembly will still be surrounded on three sides by detector banks and on the fourth side with the AmLi source. The line of detectors close to the inner wall of the cavity will have 27 tubes and the outer line 28 detectors for a PWR design (a total of 55 compared to 20 tubes in the PWR JCC-73). The BWR version will have 35 ^{3}He detectors (compared to 26 detectors for the JCC-72)

B. Photoneutron Interrogation

The Photoneutron Interrogation Device (PHONID) was developed by the JRC Ispra in 1975 (Ref. 20) to support EURATOM measurements in LEU and HEU fuel-fabrication plants. The development in the PHONID line of instruments led to the construction of four units of the PHONID 3b series (Refs. 21-23). The PHONID devices have been constructed to measure quantities of ^{235}U contained in any uranium-bearing sample by a non-destructive method. A ^{124}Sb-^{9}Be Photoneutron source performs an active interrogation of the sample with epithermal neutrons. With moderating material absent in the measuring cavity, the neutron source spectrum has an average neutron energy of 12 keV in the center of the assay chamber. The source neutrons induce fissions in the ^{235}U contained in the sample. The prompt fission neutrons are counted by ^{4}He fast neutron detectors, thereby allowing separation (by energy discrimination) of the fission neutrons from the source neutrons and γ- background. After correction for the decay of the external photoneutron source ($T_{1/2}$= 60.2 d) the neutron count rate can be related to the ^{235}U mass in the sample. The use of the PHONID as an active neutron interrogation system requires a strict stratification of the uranium waste with respect to geometry, homogeneity of the distribution and density of the fissile (absorbing) and the matrix (moderating) materials. Under these strict conditions the performance of PHONID can vary from 2% accuracy (Ref. 23) on well conditioned materials (pellets and powders).

Recently an advanced PHONID technique (Ref. 24) has been developed by the JRC Ispra in collaboration with BNFL Instruments Ltd. (UK). The technique is based on a combination of emission and transmission signals allowing 'self categorization' of samples. The measurement technique was demonstrated on LEU and HEU standards from the PERLA laboratory comprising pellets, metal, UO_2 and U_3O_8 powder ranging from 1% to 92% enrichment. Based on the emission and transmission signal, without prior knowledge of the characteristics of the sample i.e. enrichment, an overall accuracy of 2% has been obtained. The performance of the technique has also been demonstrated on scrap material originating form a HEU reprocessing facility in Europe (Ref. 23). Based on a calibration with natural uranium reference standards an accuracy of 15% can be obtained for measurements of uranium scrap samples with enrichments of up to 90%. (See Fig. 12).

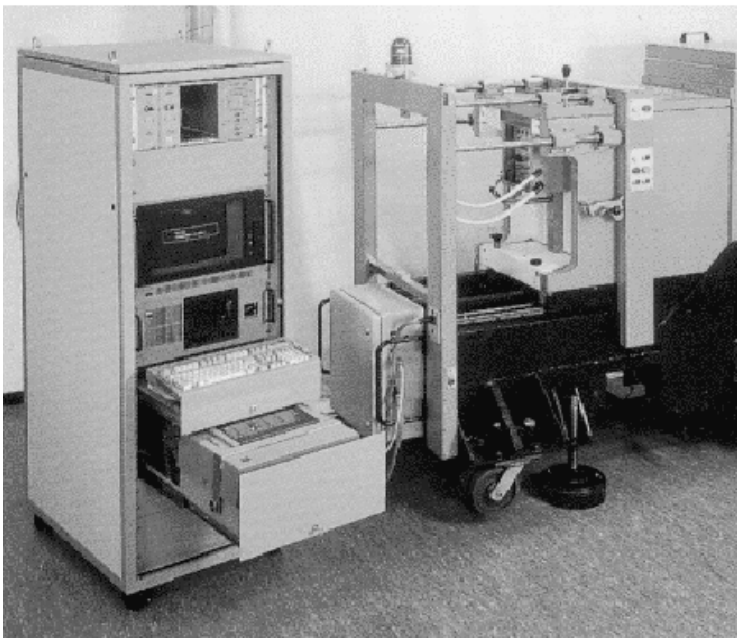

***Figure 12**: The Photoneutron Interrogation (PHONID) assay system. Data acquisition and analysis electronics are on the left, and the assay station is on the right. The massive right-hand portion of the assay station contains the photoneutron source. The left-hand portion holds the assay sample.*

C. General Comments

In table 2 the LEU fabrication facilities in Europe inspected by EURATOM are listed together with NDA techniques applied by EURATOM in these facilities. In table 3 the material safeguarded by the different instruments is given

Table 2: European LEU Fuel-fabrication facilities inspected by EURATOM

Facility	PHONID 3b	PMCA	NCC
FBFC Dessel (B)	x	x	x
FBFC Romans sur Isere (F)		x	x
ENUSA Juzbado (E)	x	x	x
BNFL Springfields (UK)		x	
ABB Atom (S)		x	x

PMCA portable multichannel analyzer system for enrichment measurements with NaI
NCC neutron coincidence collar (developed at Los Alamos). Note: Future installations of the UMS are anticipated

Table 3: Nuclear materials measured by the EURATOM NDA instrumentation.

NDA Instrument	**Material**
PHONID 3b	UO_2 and U_3O_8 powder Pellets Clean Scrap (*) Dirty Scrap (*)
Gamma spectroscopy with portable multichannel analyzer	UF_6 cylinders UO_2 and U_3O_8 powder Pellets Clean Scrap (*) Dirty Scrap (*)
NCC	Fresh Fuel assemblies

(*) Clean and Dirty scrap follow the definition from the IAEA's Safeguards Glossary (IAEA/SG/INF/1 (1987))

VI. CONCLUSIONS

Radiation-based nondestructive assay measurements provide a rapid and accurate determination of SNM content and isotopic composition in fuel-fabrication facilities. When accompanied by destructive analytical measurements, measurement control, and effective application of measurement physics, they result in timely and accurate MC&A data for better nuclear material accountancy. These NDA measurement techniques are employed by plant staff for both operations and safeguards purposes, as well as for verifications by plant, national, and international inspectors. A summary of the measurement techniques discussed and their average performance is given in Table 4.

Table 4. NDA Measurement Techniques Applied in a LEU Physical Inventory	**Isotope Assayed**	**Typical Count Time**	**Estimated Random Error (%)***	**Estimated Systematic Error for Typical Items (%)***
Low-Resolution Gamma-Ray Enrichment	^{235}U	300s	1.0	1 - 7
High-Resolution Gamma-Ray Enrichment	^{235}U	1000s	0.1	0.1 -1
High-resolution, transmission-corrected passive gamma-ray assay of solutions	^{235}U	1000s	0.3	0.1 - 2
	^{238}U	1000s	0.5 -2	1-2
Low-resolution passive gamma-ray assay of holdup deposits	^{235}U	300s	5-10	10 - 20
	^{238}U	300s	10-15	10 - 20
Segmented Gamma Scanning of Scrap & Waste	^{235}U	1000s	1.0	2 - 10
Passive Neutron Coincidence Collar	^{238}U	300s	1 - 2	2 - 4
Active Neutron Totals Counting	^{235}U	100s	0.5 - 2	2 - 10
Active Neutron Coincidence Counting	^{235}U	1000s	0.5 - 5	1 - 5
Active Neutron Multiplicity Counting	^{235}U	1000s	1 - 10	1 - 3
Neutron Coincidence Collar for Fuel Assemblies; UMS	^{235}U	1000s	0.5	2 - 4
Californium Shuffler for cans	^{235}U	1000s	0.1	0.5 - 5
Active Fuel Rod Scanner	^{235}U	30s	0.1	1.0
Photoneutron (PHONID) on product material	^{235}U	1000s	2	2
Photoneutron (PHONID) on scrap material	^{235}U	1000s	15	5 - 15

*The IAEA and ESARDA maintain extensive data-base archives of measurement performance results from all inspections.

Summarized in Table 5 are the various measurement needs in a uranium fuel-fabrication facility and the recommended NDA techniques to perform the measurements. For further detail on the information in tables 4 and 5, see Ref. 25.

Table 5. Recommended NDA techniques for uranium fuel-fabrication plant measurement needs.

Measurement Need	**Recommended NDA Measurement Approach**
• **Pellet Production [UF_6 to UO_2 conversion]**	
- Assay UF_6 feed material	
enrichment	γ spectroscopy
UF_6 weight	weighing
- Assay UO_2 product inventory	
enrichment	γ spectroscopy
fissile isotope mass	active neutron coincidence
monitored storage	gamma, neutron signatures
- Assay UO_2 pellets	
enrichment	γ spectroscopy
fissile isotope mass	active neutron coincidence
- Assay residual SNM held up in process	γ spectroscopy
• **Waste Handling**	
- Assaying high- and low-density U waste total SNM content	delayed neutron interrogation (shufflers)
- Assaying Low-density U waste	segmented gamma scanning combined with passive neutron coincidence counting
• **Fuel Fabrication**	
- Measure fuel rods for production quality & verification of SNM	
fissile isotope mass (per unit length)	rod scanner (neutron activation & delayed gamma counting)
- Measure fuel assemblies for production quality & verification of SNM	
fissile isotope mass (per unit length)	active neutron coincidence collar
• **Transportation**	
- Shipper/receiver verification measurements	γ spectroscopy, cross-calibrated neutron coincidence counters
• **Perimeter Safeguards**	
- Check personnel & vehicles for SNM	personnel portal monitors, vehicle portal monitors, scanning of personnel with hand-held instrumentation

VII. REFERENCES

1. D.R. Rogers, *ed.*, *Handbook of Nuclear Safeguards Measurement Methods*, US NRC Publication NUREG/CR-2078 (1983).

2. T.D. Reilly, N. Ensslin, and H.A. Smith, Jr., and S. Kreiner *eds.*, *Passive Nondestructive Assay of Nuclear Materials*, US NRC Publication NUREG/CR-5550 (1991).

3. T. Gozani, *Active Nondestructive Assay of Nuclear Materials*, US NRC Publication NUREG/CR-0602 (1981).

4. J. Parker, "The Use of Calibration Standards and the Correction for Sample Self-Attenuation in Gamma-Ray Nondestructive Assay," Los Alamos National Laboratory report LA-10045, Rev. (issued August 1984; revised November 1985).

5. R. Martin, D. Jones, and J. Parker, "Gamma-Ray Measurements with the Segmented Gamma Scanner," Los Alamos Scientific Laboratory report LA-7059-M (1977).

6. Parker, J.L. and Brooks, M., "Accurate, Wide-Range Uranium Enrichment Measurements by Gamma-Ray Spectroscopy," Los Alamos National Laboratory Report, LA-11277-MS (1988)

7. Smith, H.A., "Measurement of Uranium Enrichment," in *Passive Nondestructive Assay of Nuclear Materials*, N. Ensslin, T.D. Reilly, and H.A. Smith *eds*., Chapter 7 (1991).

8. L. Kull and R. Ginaven, "Guidelines for Gamma-Ray Spectroscopy Measurements of ^{235}U Enrichment," Brookhaven National Laboratory report BNL-50414 (1974).

9. Menlove, H.O., "Neutron Coincidence Instruments and Applications," in *Passive Nondestructive Assay of Nuclear Materials*, N. Ensslin, T.D. Reilly, and H.A. Smith *eds*., Chapter 17 (1991).

10. Gozani, T., "Representative Active Nondestructive Assay Systems," in *Active Nondestructive Assay of Nuclear Materials*, Chapter 9 (1981).

11. Rinard, Philip, "The ^{252}Californium Shuffler," Los Alamos National Laboratory Application Note LALP-90-15 (1990).

12. P. M. Rinard, "Shuffler Instruments for the Nondestructive Assay of Fissile Materials," Los Alamos National Laboratory report LA-12105-MS, May 1991.

13. P. M. Rinard, E. L. Adams, H. O. Menlove, and J. K. Sprinkle, Jr., "The Nondestructive Assay of 55-Gallon Drums Containing Uranium and Transuranic Waste Using Passive-Active Shufflers," Los Alamos National Laboratory report LA-12446-MS, November 1992.

14. S.T. Hsue, J.E. Stewart, P. M. Rinard, and G. Butler, "Guide to Preparation of Nondestructive Assay Calibration Standards," Los Alamos National Laboratory Report in preparation (1997).

15. H. Smith, J. Stewart, and W. Ruhter, "Design of Standards for Nondestructive Assay of Special Nuclear Material," First International Conference on Nuclear Safeguards, Obninsk, Los Alamos National Laboratory Report LA-UR-97-766 (1997).

16. Russo, P.A., *et al.*, "Generalized-Geometry Gamma-Ray Holdup Assay," Los Alamos National Laboratory Application Note LALP-94-172 (1995).

17. Ensslin, N. and Smith, H.A., "Attribute and Semi-Quantitative Measurements," in *Passive Nondestructive Assay of Nuclear Materials*, N. Ensslin, T.D. Reilly, and H.A. Smith *eds*., Chapter 20 (1991).

18. S. Guardini and J. Loeschner, “Multimedia Systems for Safeguards Training”, Proceedings of the 5th International Conference on Facility Operations-Safeguards Interface, September 24-29 1995, p. 371-377

19. Van der Eecken, M. Swinhoe, L. Becker, J. Bourdon, L. Caldon, M. Franklin, J. Gonalves, S. Guardini, J. Loeschner, S. Nonneman, F. Sorel, V. Vocino, “An Integrated System for Unattended Inspection of Fresh LEU Fuel Assemblies”, Proceedings of the 18th ESARDA Symposium Aachen, May 1996, p. 599-604

20. G. Birkhoff, L. Bondar, J. Ley, G. Busca and M. Tramontona, “^{235}U - Measurements by means of an Antimony-Beryllium Photoneutron Interrogation Device”, Internal Report- EURATOM- Ispra- 1664,1975

21. R. Bardelli, L. Becker, L. Lezzoli, W. Matthes, R. Rochez, P. Schillebeeckx, U. Weng and J.K. Sprinkle, “The Capabilities of Phonid 3b”, American Nuclear Society, The 4th International Conference on Facility Operations-Safeguards Interface, Albuquerque, September 29 - October 4, 1991

22. R. Bardelli, L. Becker, L. Lezzoli, R. Rochez, P. Schillebeeckx, U. Weng and J.K. Sprinkle, “The Measurement Capabilities of Phonid 3b”, Proceedings of the 13th Annual Symposium on Safeguards and Nuclear Material Management, Avignon, France, May 14-16 1991, ESARDA, 24, (1991), p453-457

23. S. Napier and P. Schillebeeckx, “Phonid 3b Measurements of Uranium Waste”, EUR 14551 EN, Ispra 1992

24. P. Schillebeeckx, M. Clapham, U. Graf, N. Harris, L. Lezzoli, and S. Napier, “Recent Developments in Active Neutron Interrogation of ^{235}U by Combined Transmission and Emission Measurements”, Proceedings of the 37th Annual Meeting, INMM, 28 July - 1 August 1996, Naples, Fla. (USA).

25. Ensslin, N. and Smith, H.A., “NDA Application Guide,” in *Passive Nondestructive Assay of Nuclear Materials*, N. Ensslin, T.D. Reilly, and H.A. Smith *eds*., Chapter 23 (1991).